U.S. DEPARTMENT OF COMMERCE
National Technical Information Service

PB-295 013

Flue Gas Desulfurization Pilot Study. Phase I Survey of Major Installations. Appendix 95-L Copper Oxide Flue Gas Desulfurization Process

NATO Committee on the Challenges of Modern Society, Brussels (Belgium)

Jan 79

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COMMITTEE ON THE CHALLENGES OF MODERN SOCIETY

PB 295013

Flue Gas Desulfurization Pilot Study Phase I — Survey of Major Installations - Copper Oxide Flue Gas Desulfurization Process

Appendix 95-L

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SPRINGFIELD, VA. 22161

NORTH ATLANTIC TREATY ORGANIZATION

TECHNICAL REPORT DATA

(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
95-L		PB295013

4. TITLE AND SUBTITLE	5. REPORT DATE
Flue Gas Desulfurization Pilot Study Phase I - Survey of Major Installations - Copper Oxide Flue Gas Desulfurization Process	January 1979
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) United States	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS PEDCo Environmental Cincinnati, Ohio	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS U. S. Environmental Protection Agency Office of Research and Development (RD-681) 401 M Street, S.W. Washington, D.C. 20460	13. TYPE OF REPORT AND PERIOD COVERED
	14. SPONSORING AGENCY CODE EPA/600/00

15. SUPPLEMENTARY NOTES This report is one of a series on the status of flue gas desulfurization (FGD) in the NATO countries and Japan. The United States, the Federal Republic of Germany, and Norway have acted as lead countries in gathering information and preparing of the reports.

16. ABSTRACT The copper oxide flue gas desulfurization (FGD) process is one of the few dry processes for controlling sulfur dioxide emissions. The U.S. Bureau of Mines and Shell Corporation developed this process in the early 1960's. Shell continued development and installed a pilot plant at their Permis refinery near Rotterdam in 1967. Later they installed a full-size 36-MW boiler unit at the Yokkaichi refinery in Japan. Although all problems have not been solved, the system effectively removes SO_2 from an oil-fired boiler at an efficiency of 90 percent or greater. The system has not yet been used on a full-scale coal-fired boiler, but a jointly funded U.S. Environmental Protection Agency (EPA) and Tampa Electric Co. (TECO) pilot study has just ended at TECO's Big Bend coal-fired station.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Flue gases Copper Oxide Flue Gas Desulfurization Alternatives	NATO United States	68A 97F

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
RELEASE TO PUBLIC - BEST AVAILABLE COPY	UNCLASSIFIED	57
	20. SECURITY CLASS *(This page)* UNCLASSIFIED	22. PRICE PC A04 MF A01

EPA Form 2220-1 (9-73)

NATO-CCMS STUDY

PHASE I

Flue Gas Desulfurization Pilot Study Phase I

Survey of Major Installations - Copper Oxide Flue Gas Desulfurization Process

APPENDIX 95-L

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January 1979

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SUMMARY

The copper oxide flue gas desulfurization (FGD) process is one of the few dry processes for controlling sulfur dioxide emissions. The U.S. Bureau of Mines and Shell Corporation developed this process in the early 1960's. Shell continued development and installed a pilot plant at their Pernis refinery near Rotterdam in 1967. Later they installed a full-size 36-MW boiler unit at the Yokkaichi refinery in Japan. Although all problems have not been solved, the system effectively removes SO_2 from an oil-fired boiler at an efficiency of 90 percent or greater. The system has not yet been used on a full-scale coal-fired boiler, but a jointly funded U.S. Environmental Protection Agency (EPA) and Tampa Electric Co. (TECO) pilot study has just ended at TECO's Big Bend coal-fired station.

The process employs a copper oxide (CuO) adsorbant to extract SO_2 from clean flue gas. When the CuO becomes loaded with SO_2, the flue gas is switched to a fresh reactor. Hydrogen is pumped into the loaded reactor to regenerate the CuO and release SO_2. The SO_2 off-gas is processed to liquid SO_2 or elemental sulfur.

A combination of an alternating reactor and a special parallel-passage packing design is used to overcome the adsorbant attrition and packed-bed plugging problems usually associated with a metal oxide adsorption system. The CuO adsorbant with hydrogen regeneration is particularly suited for FGD because the regeneration step occurs at the same rate or faster than the adsorption step, and both occur at the same temperature.

The copper oxide system is currently being investigated for its potential as a viable combined SO_2 and NO_x removal system. The simple addition of ammonia to the reactor causes the CuO to act as a catalyst, removing up to 70 percent NO_x.

The copper oxide FGD system is applicable to new boiler systems located in industrialized areas where a market for sulfur based by-products (S, SO_2, H_2SO_4) exist, land is at a premium, hydrogen is available, and where the potential for NO_x control exists. This system has achieved 90 percent SO_2 control, but has not operated on a full-scale basis on a coal-fired boiler.

The following are advantages and disadvantages of the copper oxide system compared with other FGD systems:

Advantages

1. Since the system uses a dry process, wet materials are not handled, and water input requirements are minimal.

2. Acceptance and regeneration occur at approximately the same temperature, obviating any heating or cooling of the absorption beds.

3. The flue gas passes over the surface of the acceptor material rather than through it. Plugging of the acceptor bed does not occur.

4. By alternating the acceptor and regeneration units, continuous processing can be maintained.

5. Operating costs are reduced, since flue gas reheating is unnecessary and water requirements are low.

6. The process has the potential for conversion to a combined SO_2 and NO_x removal system by ammonia injection into the acceptor bed. The CuO will act as a catalyst for NO_x absorption.

7. The reactor can be operated either upstream or downstream of an electrostatic precipitator.

Disadvantages/Problems

1. Equipment and installation costs are high.

2. A hydrogen source is needed for regeneration, and the large hydrogen requirements may result in high raw material costs.

3. Damper operation becomes critical when operational modes are switched at high temperature.

4. The stripper uses larger quantities of low pressure steam, thus requiring high energy input.

5. The high temperature (400°C) required for operation may make installation on existing boilers difficult. The system can be installed between the boiler and air preheater in new plants.

1.0 BACKGROUND AND INTRODUCTION

This report is one of a series on the status of flue gas desulfurization (FGD) in the NATO countries and Japan. The United States, the Federal Republic of Germany, and Norway have acted as lead countries in gathering information and preparing of the reports.

The Shell Corporation and the Bureau of Mines began development of the copper oxide FGD process in the early 1960's. The objective was to develop a process using a dry selective adsorbant to avoid complications characteristic of wet systems. A further objective was to keep energy requirements low. Early work concentrated on using CuO sorbent granules precipitated from solution or a CuO powder. When this approach proved unsuccessful, efforts were directed toward fixing copper in a strong porous support. Alumina and macroporous silica gave the best results. Shell investigated various types of reactors, but found that a parallel-passage reactor with isolated regeneration cycling between two reactors had the most advantages.

In 1967 Shell began demonstrating the copper oxide system on a slipstream (670 to 1000 Nm^3/h) from a high-sulfur fuel-oil process heater at their Pernis refinery near

Rotterdam in the Netherlands. Ninety percent SO_2 removal was achieved over a period of 20,000 operating hours. Sorbent life was estimated at 1.5 years.

A commercial unit was installed in 1973 at the Showa Yokkaichi Sekiyu (SYS) oil-fired 40-MW boiler (125,000 Nm^3/h and 2500 ppm SO_2). Ninety percent SO_2 removal was achieved. The SO_2 was processed in a Claus plant with normal refinery acid gas and also in an absorber stripper. By injecting ammonia, NO_x was removed simultaneously at an efficiency up to 70 percent.

To test this process on a coal-fired power plant, the U.S. Environmental Protection Agency (EPA) and Tampa Electric Co. (TECO) have jointly funded a demonstration plant on a slipstream of TECO's Big Bend Station.

2.0 GENERAL PROCESS DESCRIPTION

The copper oxide FGD process employs a dry acceptor, CuO on alumina, in a fixed packed-bed reactor, to extract sulfur oxides from flue gas containing oxygen and particulate matter. Desulfurization efficiency decreases as the acceptor becomes loaded with sulfur. The material is regenerated in situ and at the same temperature at which it was accepted to release the sulfur as sulfur dioxide in the regeneration off-gas. Two or more identical reactors are applied in swing operation to provide continuous processing of flue gas. The SO_2 in the off-gas from regeneration, free from oxygen and particulate matter, can be processed to yield liquid SO_2 or elemental sulfur. Figure 1 presents a flowsheet of a typical Shell/Universal Oil Products system as it might be applied to a 500-MW coal-fired boiler.

2.1 PROCESS CHEMISTRY

Cupric oxide was chosen among various metal oxides because CuO readily reacts with SO_2 in the presence of O_2 at 400°C to yield $CuSO_4$. The $CuSO_4$ can easily be reduced at the same temperature to yield SO_2; $CuSO_4$ is also stable to thermal dissociation at the acceptance/regeneration temperature.

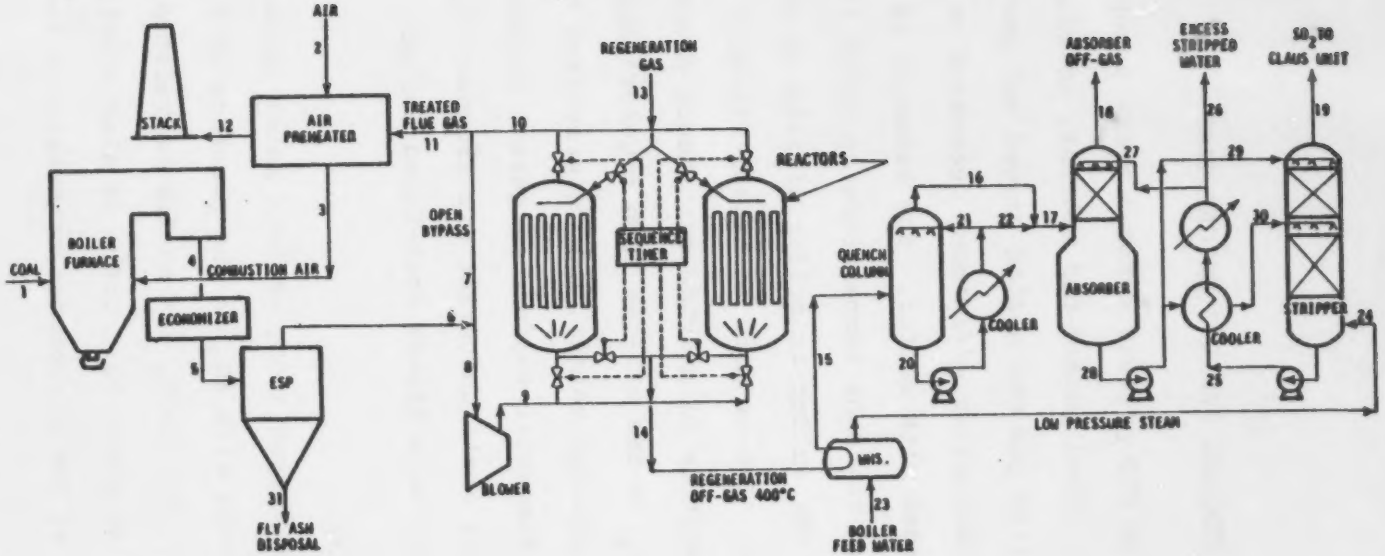

Figure 1. Flow diagram and material balance for the copper oxide process as applied to a 500-MW coal-fired boiler.

(continued)

Stream no.	1	2	3	4	5	6	7	8	9	10	11
Description	Coal to boiler	Air to preheater	Air to boiler	Gas to economizer	Gas to ESP	Gas to blower	Gas bypass to blower	Combined gas to blower	Gas to reactor	Off-gas from reactor	Gas to air heater
Rate, 1000 kg/h	170	2049	1848	1983	1998	1983	314	2297	2297	2281	1967
m^3/s		464	419	445	445	445	71	516	516	514	443
m^3/s, Liquid											
Particulates, kg/h				19,300	15,300	103	16.5	118.5	118.5	118.5	103.0
Temperature, °C		43.3	279			488	488	488	488	488	488
Sulfur as SO_2, kg/h	11,900			11,900	11,900	11,900	190	12,090	12,090	1388	1190

Stream no.	12	13	14	15	16	17	18	19	20	21
Description	Gas to stack	Regeneration gas to reactor	Off-gas from reactor	Gas to quench column	Gas to absorber	Combined gas to absorber	Absorber off-gas	Stripper off-gas	Quench column underflow	Recycle to quench column
Rate, 1000 kg/h	2160	5.355	21.4	21.4		21.4				
m^3/s	488	4.81	5.85	5.85						
m^3/s, Liquid										
Particulates, kg/h	103.0	-0-	-0-	-0-	-0-	-0-	-0-	-0-	-0-	-0-
Temperature, °C		260	488	110						
Sulfur as SO_2, kg/h	1190	-0-	10,710	10,710		10,710	-0-	10,700		

Stream no.	22	23	24	25	26	27	28	29	30	31
Description	Recycle to absorber	Water to water boiler	L.P. steam from boiler	Stripped water	Excess stripper water	Stripped to water absorber	Spent water from absorber	Spent water to stripper	Spent water to stripper	Fly ash disposal
Rate, 1000 kg/h					240					
m^3/s										
m^3/s, Liquid										
Particulates, kg/h	-0-	-0-	-0-	-0-	-0-	-0-	-0-	-0-	-0-	15.2
Temperature, °C		-0-	-0-		10					
Sulfur as SO_2, kg/h										

Notes:

1 Calculations based on:

a. 3.5% sulfur in coal (dry).
b. 13% ash coal (as fired).
c. 92% of sulfur in coal evolves as SO_2.
d. 75% of ash in coal evolves as fly ash.
e. 99.5% removal of particulates to scrubber.
f. 90% SO_2 removal.

* Particulates should be added to get total stream rate.

Figure 1 (continued).

The process is easily divided into three reaction steps: oxidation, acceptance, and regeneration.

2.1.1 Oxidation

The copper in the acceptor is present primarily as elemental copper, but small quantities occur as various copper sulfides. Upon contact with oxygen in the flue gas, the copper and any Cu_2S and CuS react rapidly to form CuO, $CuSO_4$, and SO_2:

$$Cu + 1/2\ O_2 \rightarrow CuO$$

$$Cu_2S + 5/2\ O_2 \rightarrow CuO + CuSO_4$$

$$CuS + 3/2\ O_2 \rightarrow CuO + SO_2$$

The formation of Cu_2S is undesirable because one-half the Cu_2S is converted to $CuSO_4$ during subsequent oxidation and therefore becomes unavailable for reaction with SO_2.

2.1.2 Acceptance

SO_2, O_2, and CuO react to form $CuSO_4$:

$$CuO + 1/2\ O_2 + SO_2 \rightarrow CuSO_4$$

The SO_2 is reacted until the conversion of the CuO to $CuSO_4$ has proceeded to the extent that the unconverted CuO reaches a low level and the SO_2 that escapes capture reaches the maximum acceptable limit. The loaded acceptor is then regenerated. This acceptance reaction is shown graphically in Figure 2 by expressing the SO_2 content of the treated gas as a function of elapsed time after flue gas is contacted

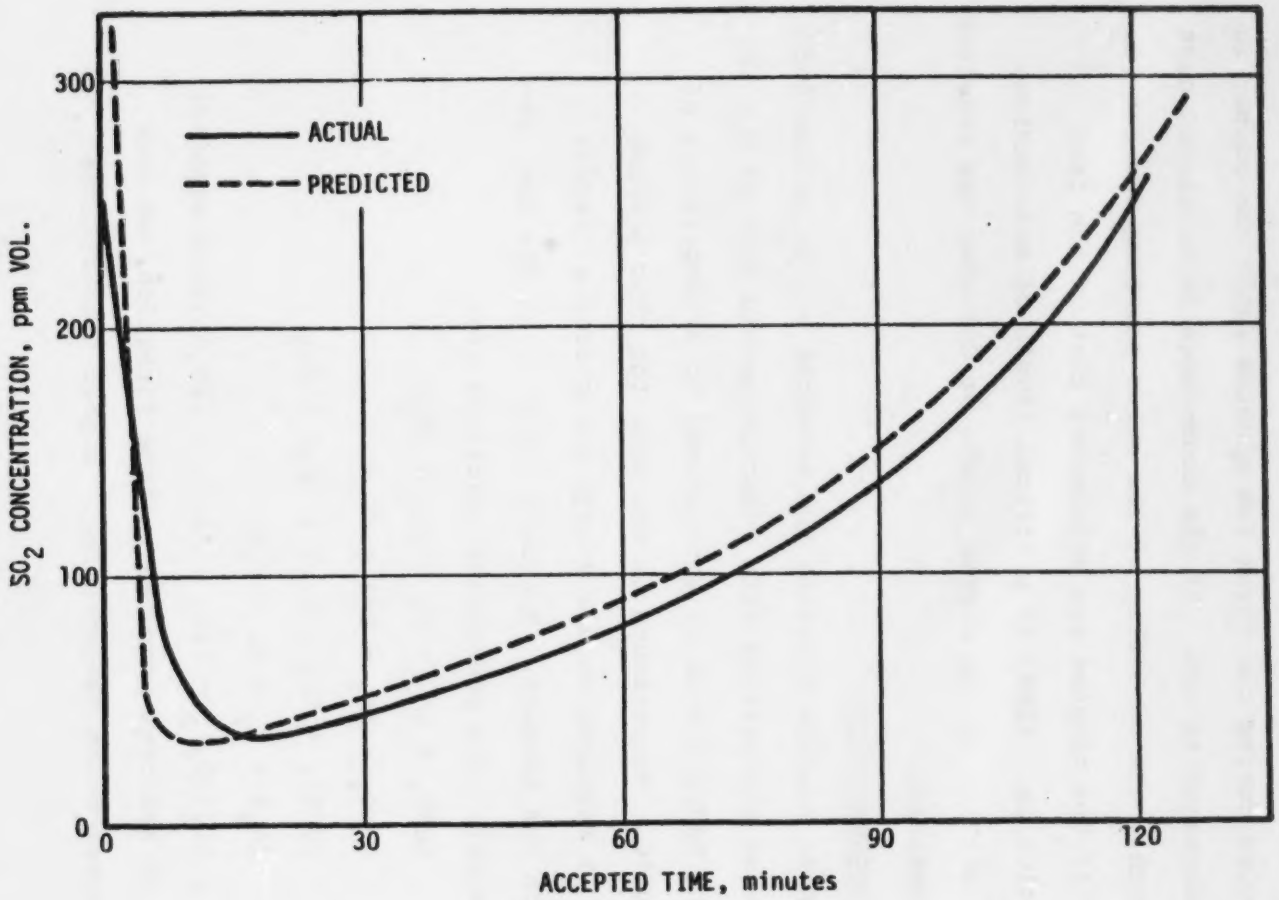

Figure 2. SO_2 concentrations in treated flue gas.

with the regenerated acceptor. The high SO_2 level at the beginning of the cycle represents the low SO_2 reaction rate that occurs during the first few minutes while the copper is being converted to CuO. If the acceptance is continued past 120 minutes, the concavity of the curve reverses and the SO_2 content of the treated gas approaches that of the feed asymptotically. Thus, at practical levels of desulfurization, a portion of the copper exists as CuO when the acceptor is regenerating.

2.1.3 Regeneration

Copper sulfate releases the accepted sulfur in the form of SO_2 when regenerated with reducing agents such as H_2, CO, and light hydrocarbons or when raised to a temperature of about 700°C. Temperature is not used for this purpose because it requires excess energy and places a thermal stress on the sorbent. Hydrogen seems to be the best reducing agent. The postulated reactions are:

$$CuSO_4 + 2H_2 \rightarrow Cu + SO_2 + 2H_2O$$

$$CuSO_4 + 4H_2 \rightarrow CuS + 4H_2O$$

$$2CuSO_4 + 6H_2 \rightarrow Cu_2S + 6H_2O + SO_2$$

$$Cu_2S + H_2 \rightarrow Cu + H_2S$$

$$Al_2(SO_4)_3 + 12H_2 \rightarrow Al_2S_3 + 12H_2O \text{ (from alumina)}$$

The use of hydrogen promotes sulfide formation, as does lower regeneration temperatures. Higher temperatures

promote $Al_2(SO_4)_3$ reduction. Reaction with SO_3 causes $Al_2(SO_4)_3$ to form in the alumina support material. Close control of temperature to approximately 400°C is required to reduce the H_2 consumption and to better reactivate the copper. Another advantage of using H_2 is that regeneration can occur at the same temperature as the oxidation and acceptance step.

The use of methane reduces sulfide formation, but it increases either the time or the temperature of regeneration. The reaction postulated is shown below:

$$CuSO_4 + 1/2\ CH_4 \rightarrow Cu + SO_2 + 1/2\ CO_2 + H_2O$$

Higher hydrocarbons reduce the $CuSO_4$ acceptably, but tend to leave coke-like deposits on the sorbent. These are readily burned off during the oxidation step; however, they tend to cause high temperatures, which increase temperature control problems and thermal stress.

Depending on the exact reactions involved, 2 or 4 mol of H_2 are required to reduce the $CuSO_4$ to Cu and CuS, respectively. Assuming 25 percent excess H_2 is required, the estimated average H_2 requirement is approximately

$$\frac{3 \times 2}{64} \times 1.25 = 0.117 \text{ kg } H_2/\text{kg } SO_2 \text{ removed}$$

Actual experience has shown H_2 requirements of 0.125 kg H_2/kg SO_2 removed.

2.1.4 Absorber Stripper

After regeneration, the SO_2 and water vapor pass through a waste heat boiler, a quench tower, and finally to the absorber stripper section where the following simplified reactions occur:

Absorber

$$SO_2 + H_2O \rightarrow H_2SO_3(aq)$$

Stripper

$$H_2SO_3 \overset{\text{Heat}}{\rightleftharpoons} SO_2 + H_2O$$

This section also serves to level out the fluctuations of SO_2 production from the reactor.

2.1.5 End Product

The end product choice is liquid SO_2 or elemental sulfur, which is produced by the Claus process. Liquid SO_2 is made by drying, compressing, and then condensing the gas from the stripper. The resulting liquid SO_2 is collected and stored in pressurized vessels.

Elemental sulfur is recovered in two steps. In the reduction section, a portion of the SO_2 reacts with methane and produces a mixture of elemental sulfur, hydrogen sulfide, carbon dioxide, water vapor, and unreacted SO_2.

$$2CH_4 + 3SO_2 \rightarrow 2CO_2 + 2H_2O + S + 2H_2S$$

In the Claus section of the process, which follows, the hydrogen sulfide and remaining SO_2 from the reduction

section react to produce additional elemental sulfur and water vapor:

$$2H_2S + SO_2 \rightarrow 2H_2O + 3S$$

The tail gas is incinerated and recycled to the reactor.

2.2 DESCRIPTION OF EQUIPMENT COMPONENTS

A copper oxide FGD unit may be applied to a boiler in either of two ways. It can be integrated between the economizer and air preheater to obtain the temperature required for the acceptance reaction, or it can be installed as an add-on downstream from the air preheater. The latter application requires additional fuel to increase the gas temperature to 400°C and a heat exchanger to recover some portion of the added thermal energy.

2.2.1 Process Particulate Removal

Particulate loading of flue gas from oil-fired boilers normally ranges from 50 to 300 mg/Nm^3, which is low enough not to interfere with reactor operation. If space allows, or if the system is newly designed, the flue gases are removed after they are treated in the economizer and then returned to the boiler preheater.

A coal-fired boiler presents a different problem. Because particulate loading of the flue gas is normally 50 to 100 times the 300 mg/Nm^3 acceptable limit of the reactor, an electrostatic precipitator (ESP) must be installed before

the reactor. (New evidence from the EPA-TECO demonstration project suggests that the reactor will not plug if it is placed before the ESP, and the abrasive nature of the fly ash may actually scour the reactor of any deposits.) In a retrofit situation an ESP is normally already in place after the preheater. The flue gas must be heated to 400°C before it enters the reactor. The cleaned gas should then be passed through a heat recovery system before venting to the atmosphere. In a newly designed system, a hot ESP and the reactor can be integrated between the economizer and the preheater.

2.2.2 Reactor

The reactor in the copper oxide system consists of two parallel flow units that alternately adsorb and regenerate i.e., while one is adsorbing the other is regenerating. The purpose of this design is to allow continuous operation and overcome sorbent attrition. Tests indicate that when a pelletized sorbent is in a moving bed or fluidized bed, agitation will destroy sorbent. In a parallel passage packing adsorber, the flue gas flows in channels parallel to beds of sorbent. The sorbent is held in thin sheets to present a large surface area, a design that alleviates the dust accumulation problem that would occur in a normal packed bed design, where the sorbent bed is perpendicular to

the gas flow. The reactor at the Yokkaichi refinery is modular and 68 unit cells are used to control the 36-MW output of the boilers. The unit cell design permits a scale-up to larger plants simply by adding more cells in parallel. Adding cells in series increases SO_2 removal efficiency. Replacement of unit cells, maintenance, and inspection are all relatively easy.

The reactor body is made of a chrome-molybdenum steel. The sorbent is CuO supported by activated alumina held in thin sheets by a fine stainless steel gauze.

The flue gas is switched from the "full" reactor (loaded with SO_2) to the freshly regenerated reactor by an automatic sequence timer. The sequence timer determines when the reactor is full, switches reactors, and then sets the regeneration sequence in motion. Because the regeneration reaction uses H_2 and the acceptance reaction contains O_2 at high temperatures, an explosive mixture could be formed. Three steps are taken to prevent such an occurrence: (1) the reactor is purged with steam at 400°C; (2) special precautions are taken to use gas valves and dampers that provide a 100 percent seal (e.g., a continuous purge of the valve seal with high-pressure steam); and (3) explosion limit calculations are made on the gases present to assure that an explosive mixture will not occur under normal operating conditions.

The cleaned flue gases pass out of the reactor to the air preheater. The air preheater is able to extract more heat from the flue gases than normal because most of the SO_3 is removed, thereby reducing the dew point of the flue gas. This prevents visual emission limits from being exceeded and enables flue gas to be released at a much lower temperature.

The off-gas from regeneration, which contains SO_2, water vapor, some H_2, and inerts, is forced through a waste heat boiler to remove usable heat and then to a quench tower to cool it even further in preparation for the absorber stripper section.

2.2.3. Absorber Stripper

The absorber stripper is used at the Yokkaichi refinery to reduce fluctuations of SO_2 gas and separate SO_2 from water vapor so that the Claus conversion is not suppressed. The flow fluctuations of the stripper overhead are reduced by incorporating surge capacity in the bottom of the absorber. Liquid pumped from the stripper contains SO_2 concentrations of 20 to 50 ppm, 75 percent as sulfates. This water could be used as boiler feed water after it is processed or, because of its small quantity, be released to the atmosphere in the flue gas.

Because the absorber stripper method of processing the SO_2-rich off-gas is very inefficient, other systems of processing are being developed.

2.3 OPERATING PARAMETER

The operating temperature of the reactor is 400°C and the optimum copper content of the acceptor is from 4 to 6 percent. A 500-MW unit treating gas containing 2000 ppm SO_2 would require acceptor and regenerator volumes ranging from 50 to 100 m^3 each. About 50 percent of the off-gas from the regenerator is SO_2. Pressure drop across the reactor is about 2100 Pa.

The absorber stripper section has an efficiency of 99.9 percent. The total sulfur content of the stripped water is 20 ppm, and the wastewater contains very low impurities. Overall pressure drop for the system ranges from 4500 to 5600 Pa.

3.0 CURRENT STATUS, ASSESSMENT, AND PLANS

At present only one oil-fired boiler in the world is operating with a full-scale copper oxide FGD system. (Tables 1 and 2 summarize the status of this system.) The system has been studied in three pilot plants, two of which were for the purpose of ascertaining the effects of coal combustion gases on the system.

The full-scale operation (at the Yokkaichi refinery in Japan) has shown an SO_2 removal rate of 90 percent, but high availability has not been achieved. The pilot plant at the Galileistraat Power Station in Rotterdam in the Netherlands demonstrated the effectiveness of the parallel passage principle in coping with high particulate loadings from a coal-fired utility boiler and suggested that the system could function successfully through oxidation-reduction cycles. The results of the jointly funded EPA-TECO pilot study at TECO's Big Bend power station will have a big impact on the future of copper oxide systems in the United States.

The copper oxide FGD process is applicable to new boilers located in industrialized areas where a market

Table 1. SUMMARY OF STATUS OF SURVEYED FGD SYSTEMS

All Systems	
Total number of systems in operation by Dec. 1977	1
Total number of systems built	1
Total number of systems surveyed	1
Year first placed in commercial service	1973
Number of utility systems using process	0
Total MW of these facilities	0
Number of industrial combustion systems	1
Surveyed Systems	
Range of average SO_2 removal efficiencies, %	80-90
Range of fly ash removal efficiency, %	0
Range of average availability factors[a], %	N.A.

[a] Availability factor computation in Appendix B.

N.A. - Not available.

Table 2. STATUS OF SURVEYED FGD INSTALLATIONS -

COPPER OXIDE PROCESS

Name of plant	Location	Size, MW	Fuel type	Characteristics		Flue gas treated		Year FGD systems started	SO_2 removal efficiency, %	Disposition of end products	Energy required % total input	Average % availability
				% S	Heat value, kcal/kg	Nm^3/sec	% of total					
Yokkaichi Refinery Showa Yokkaichi Sekiyu Co. Ltd.	Yokkaichi City, Mie Pref. Japan	36	Heavy oil C	2.5	9700	32	100	1973	85	Elemental sulfur is sold	2.4 Reheat not necessary	Unit is used as a prototype

exists for the sulfur based by-products, land is at a premium, and an inexpensive hydrogen source is available. Control of SO_2 above 95 percent appears to be a problem inherent in the cyclical nature of the system. The system is highly developed, but as yet has not demonstrated continued full-scale operation on coal-fired boilers. Effective NO_x control has been obtained with the addition of ammonia gas. It is an attractive system for limited application, but has a high capital cost.

The future of the copper oxide process is difficult to predict. No new installations are presently planned, but the system could become a good choice if its potential for simultaneous NO_x removal were developed and regulations made such removal necessary.

4.0 RECENT TECHNOLOGICAL DEVELOPMENTS AND SOLUTIONS TO PROBLEM AREAS

4.1 NEW DEVELOPMENTS - NO_x REMOVAL

The Shell copper oxide process at the Yokkaichi refinery is reported to have an NO_x removal efficiency as high as 70 percent. Ammonia added to flue gas reacts with NO at the reactor temperature to form nitrogen and water. The ammonia concentration in the flue gas released to the atmosphere is 2 ppm.

The study recently completed at TECO's Big Bend Station shows that the commercial acceptor manufactured by Ketjen has been demonstrated to be chemically and physically stable. The parallel passage reactor is an acceptable design for flue gas fly ash loadings as high as 25 g/Nm^3. Pressure drop and SO_2 removal were unaffected after 2210 acceptance-regeneration cycles over 3 months. In case they were found to be necessary, in situ cleaning techniques were developed. It was determined that it is not necessary to have fly ash removal upstream of the CuO reactor when used on a coal fired boiler.

4.2 SOLUTION TO PROBLEM AREAS

The Yokkaichi refinery has experienced three problems: (1) dust deposition around the inlet of the reactor, (2) plugging of the waste heat boiler, and (3) corrosion of the quenchtowers. The first problem was solved by installing steam soot blowers where the deposits collect. Plugging of the waste heat boiler and corrosion of the quenchtower are expected to be solved by changing the material and the structure of these pieces of equipment.

5.0 COST

Cost data for the copper oxide process are very limited, since there is only one operating plant. Costs reported in the single questionnaire were assumed to apply to the date of plant start-up unless otherwise indicated. The cost at initiation of construction was converted to January 1976 dollars using the Chemical Engineering plant cost index (1957-59 = 100 and Jan. 1976 = 187.1). The adjusted costs are presented in Table 3 and actual costs in the Appendix.

The Yokkaichi refinery reported only capital costs for 1973. This refinery reports that one man-year of specialist-level labor is necessary. If $10/h is assumed for this labor, and 15 percent is added for supervision, annual labor costs amount to $100,000. If $0.38/kg of hydrogen is assumed for the raw materials and 218 Mg/yr are needed, annual raw material costs amount to $85,000. Maintenance is estimated at 4.35 percent of total capital investment and 15 percent of labor and materials, and results in an annual maintenance cost of $191,000.

Cost estimates have been published based on the recently completed study at TECO's Big Bend Station. The cost

Table 3. COST DATA OF INDIVIDUAL INSTALLATIONS FOR THE COPPER OXIDE FGD SYSTEM

Plant name Location	Yokkaichi Refinery Yokkaichi City
1. Reported capital costs, $	3,330,000
Year	1973
Cost inflated to 1976	4,712,400[a]
$/kW	130.9
2. Reported annual costs, $	
Total fixed costs	878,862[b]
Fixed charge factor, %	18.65
Total variable costs	N.A.
3. Boiler operating hours	8040
Capacity factor	0.90
Generated MWh (equivalent)	283,824
4. Reported cost per kWh, mills/kWh	1.69-4.24[c]
Fixed cost	3.10
Variable costs	N.A.
Total cost, mills/kWh[d]	
4000 h	
8000 h	

[a] Fixed costs are normally based on 7 years' depreciation in Japan. This estimate is based on 20 years.

[b] Yen is converted to dollars in year of construction initiation and then converted to January 1976 dollars using the Chemical Engineering Plant Cost Index (1957-59 = 100, 1976 = 18[illegible], 1 dollar = 300 yen).

[c] Operating cost estimated by Shell.

[d] Based on capacity factor of 100 percent.

N.A. - Not available.

study was patterned after that used by the Tennessee Valley Authority (TVA) in a previous study for the EPA. The TECO study provided estimated capital and operating costs for mid-1977 for 90 percent desulfurization of the flue gas from a new 500-MW, coal-fired boiler and are shown in Table 4. Sensitivity to sulfur content was shown with three sulfur levels: 3.5, 2.5 and 0.8 percent.

Table 4. Capital and Operating Cost of CuO Systems

Cost estimate	1	2	3
Sulfur content, %	3.5	2.5	0.8
Capital cost, $/kW	97	88	62
Operating cost, mills/kWh	3.8	3.2	1.9

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APPENDIX A

APPENDIX A

QUESTIONNAIRE DATA ON THE COPPER OXIDE FLUE GAS DESULFURIZATION PROCESS

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
1.	GENERAL DESCRIPTION	
1.1	Company	Showa Yokkaichi Sekiyu Co. Ltd. (SYS)
1.2	Plant name	Nihon Shell Gijutsu/SIPM NGC Corp.
1.3	Preparerb name and mailing address	Jumpei Ando
1.4	Principal plant products	Petroleum products
	Brief description of area surrounding plant site and factors for choice of FGD system	Located near Yokkaichi city; since a serious air pollution problem 10 years ago, stringent SO_2 emission regulations have been applied. Four boilers use low-sulfur oil. One uses FGD in conjunction with Claus process to recover elemental sulfur.
1.5	Plant location	Yokkaichi City, Mie Prefecture
1.6	STATION CAPACITY	
1.6.1	Total boiler generating capacity MW / number of boilers	110/5
1.6.2	Capacity served by FGD system, MW	36
	Number of boilers served	1
	% of boiler flue gas treated by FGD system	100

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
1.7	TYPE OF FGD SYSTEM	CuO absorption (dry process)
1.8	Number of scrubber trains in parallel	2 (acceptor and regenerator, switching).
1.9	FGD system installed on new or existing boiler	Existing
1.10	STATUS OF FGD SYSTEM operational or under construction, date started	Operational, June, 1973
2.	BOILER DATA	
2.1	Boiler identification number	No. 6
2.2	Maximum continuous heat input, MW	87.2
2.3	Max. cont. net generating capacity, MW	40
2.4	Flue gas flow rate, Nm^3/h temperature at induced draft fan, °C	115,000 N.A.
2.5	Boiler manufacturer	N.A.
2.5.1	Fuel type	Heavy oil, grade C
2.5.2	Firing configuration	Front firing, 6 burners
2.6	Year placed in service	1972
2.7	Type service	Base load
2.8	Stack height, m	110
2.8.1	Flue gas exit velocity, m/s	25.8
2.9	Average boiler operation, h/yr	8040
2.10	Average boiler capacity factor	0.90

N.A.- Not available.

(continued)

APPENDIX A (continued)

	NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
2.11 COAL COMPOSITION	None
2.11.1 Lower heating value, kJ/kg	
2.12.2 Sulfur content , %	
2.11.3 Ash content , %	
2.11.4 Chloride content , %	
2.11.5 Moisture , %	
2.12 OIL COMPOSITION	
2.12.1 Grade/heating value , kJ/kg	Heavy oil/ 40,610
2.12.2 Sulfur content , %	2.5
2.12.3 Ash content , %	0.018
2.12.4 Chloride content , %	N.A.
2.13 PARTICULATE REMOVAL EQUIPMENT NOT PART OF FGD SYSTEM	N.A.
2.13.1 Type	
2.13.2 Manufacturer	
2.13.3 Efficiency, % removal	
2.13.4 Emission rate , mg/Nm^3	

(continued)

APPENDIX A (continued)

	NIHON SHELL GIJUTSU/SIPH NGC CORP. Yokkaichi Refinery
3. EMISSION REGULATIONS	
3.1 ATHOSPHERIC EMISSIONS	
3.1.1 SO_2, Nm^3/h	64.8
3.1.2 Particulate, mg/Nm^3	50
3.1.3 NO_x, ppm	230
3.1.4 Other	N.A.
3.1.5 Legal basis	by HIE Prefecture
3.2 RESTRICTIONS APPLICABLE TO LIQUID OR SOLID WASTES, LEGAL BASIS	N.A.
4. FGD SYSTEM INFORMATION	
4.1 Process name	Shell copper oxide process
4.1.1 Supplier or design company	Nihon Shell Gijutsu/SIPH NGC Corp.
4.1.2 WHY WAS THIS SYSTEM CHOSEN?	Concentrated SO_2 can be recovered for conversion to elemental sulfur by existing Claus process.
4.1.3 PROCESS DESCRIPTION	Appendix Figure A

(continued)

APPENDIX A (continued)

	NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
4.2 SO_2 SCRUBBING AGENT	
4.2.1 Name	SFGD Acceptor
4.2.2 Chemical composition	CuO - Activated Al_2O_3
Components, % by weight	CuO, approximately 5
4.2.3 Stoichiometric requirement	N.A.
4.2.4 Addition rate, kg/h	N.A.
4.2.5 Particle size, mm	1.2 (diam.), 3 (length)
4.3 REGENERATION AGENT	
4.3.1 Name	Reducing gas (diluted)
4.3.2 Composition	H_2 with some CH_4 (20%) and steam (balance)
4.3.3 Stoichiometric requirement	2.5-4.0 mole H_2/mole SO_2
4.3.4 Addition rate	N.A.
4.3.5 Particle size	N.A.
4.4 SALABLE PRODUCT	
4.4.1 Name	Sulfur
4.4.2 Chemical composition Components,% by weight	S,99.9
4.4.3 Amount generated/year, Mg/yr	1100
4.4.4 Sales potential	All has been sold.

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM HGC CORP. Yokkaichi Refinery
4.4.5	BY-PRODUCT RECOVERY PROCESS	
	Name	Claus furnace
	Vendor	Mitsubishi Kakoki K.K.
	Process description	N.A.
4.4a	Intermediate product composition	Concentrated SO_2 Gas
4.5	BY-PRODUCT NO. 2	None
4.6	WASTE STREAMS	N.A.
4.6.1	Type of waste	
	% water	
4.6.2	Amount generated in 1 hour at full load	
4.6.3	Disposal method	
4.6.4	Disposal site and distance from plant, km	
4.6.5	Potential effects on environment from sludge disposal	
4.7	SLUDGE FIXATION PROCESS	N.A.

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM NCC CORP. Yokkaichi Refinery
4.8	FGD EQUIPMENT DATA	None
4.8.1	FIRST SCRUBBER	
4.8.1.1	Type	
4.8.1.2	L/G[a] ratio, ℓ/Nm^3	
4.8.1.3	Gas velocity through absorber or scrubber, m/s	
4.8.1.4	Absorbent (adsorbent) flow, m^3/h	
4.8.1.5	Gas flow entering scrubber, Nm^3/h	
4.8.1.6	Gas temperature, °C	
4.8.1.7	Gas composition	
	SO_2 in → out, ppm	
	% removal	
	Particulate in → out, mg/Nm^3	
	% removal	
	NO_x in → out, ppm	
	CO_2 in → out, ppm	
	Other in → out	
4.8.1.8	Material of construction	
4.8.1.9	Type of lining	
4.8.1.10	Absorbent and sand losses	
4.8.1.11	Number of stages	
4.8.1.12	Packing type and composition	

[a] Liquid-to-gas ratio

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPH NGC CORP. Yokkaichi Refinery
4.8.2	SECOND SCRUBBER	
4.8.2.1	Type	Parallel passage reactor
4.8.2.2	L/G ratio, ℓ/Nm^3	N.A.
4.8.2.3	Gas velocity through absorber or scrubber, m/s	N.A.
4.8.2.4	Absorbent (adsorbent) flow, m^3/h	N.A.
4.8.2.5	Gas flow entering scrubber, Nm^3/h	110,000
4.8.2.6	Gas temperature, °C	400
4.8.2.7	Gas composition	
	SO_2 in + out, ppm	1250 + 125-250
	% removal	80-90
	Particulate in + out, mg/Nm^3	Below 50 + Below 50
	% removal	
	NO_x in + out, ppm	330 + Below 200
	% removal	Over 40
	CO_2	N.A.
	Other	N.A.
4.8.2.8	Material of construction	1-1/4 Cr - 1/2 Mo
4.8.2.9	Type of lining	N.A.
4.8.2.10	Number of stages	N.A.
4.8.2.11	Packing type and composition	N.A.

(continued)

APPENDIX A (continued)

		NIHON SHALL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
4.8.3	CLEAR WATER TRAY	N.A.
4.8.3.1	Type	
4.8.3.2	L/G ratio, ℓ/Nm^3	
4.8.3.3	Source of water	
4.8.4	DEMISTER	N.A.
4.8.4.1	Type	
4.8.4.2	Number of passes (stages)	
4.8.4.3	Material of construction	
4.8.4.4	Method of cleaning	
4.8.4.5	Flow rate during cleanings, Nm^3/h	
4.8.4.6	Frequency and duration of routine cleaning	
4.8.5	REHEATER	N.A.
4.8.5.1	Type	
4.8.5.2	Heat load, kW	
4.8.5.3	Heat transfer surface area, m^2	
4.8.5.4	Temperature of gas in, °C out, °C	
4.8.5.5	Reheater materials of construction	

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
4.8.6	SCRUBBER TRAIN PRESSURE DROP, mbar	
4.8.6.1	First scrubber	N.A.
4.8.6.2	Second scrubber	Reactor 19.6
4.8.6.3	Clear water tray	N.A
4.8.6.4	Demister	N.A.
4.8.6.5	Reheater	N.A
4.8.6.6	Ductwork	N.A.
4.8.6.7	Total FGD system	39.2-49.0
4.8.6.8	I.D. fan	N.A.
4.9	SYSTEM MAKEUP WATER ADDED, m^3/h	N.A.
4.9.1	Water added per mole of SO_2 removed, m^3/kg mol	
4.9.2	Source of makeup water	
4.9.3	Where is water added	
4.10	BYPASS SYSTEM	
4.10.1	Can flue gas be bypassed?	Yes
4.10.2	Height of gas discharge when bypassed, m	110
4.11	SLURRY DATA, pH, % solids by weight	N.A.
4.11.1	Fresh slurry to scrubber	
4.11.2	Recirculating slurry	
4.11.3	Waste slurry	

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
4.12	SPENT SLURRY SYSTEM	N.A.
4.12.1	Clarifiers	
4.12.1.1	Number	
4.12.1.2	Diameter, m	
4.12.2	Solid separation	
4.12.2.1	Type	
4.12.2.2	Cloth area, m^2	
4.12.2.3	Capacity (wet cake), ton/h	
4.13	SPACE REQUIRED, EXCLUSIVE OF SLUDGE DISPOSAL AREA, m^2	600 (excluding Claus furnace)
4.14	PERSONNEL, man-hours/yr	
4.14.1	Operating	N.A.
4.14.1.1	Specialists	1
4.14.1.2	Unskilled	0
4.14.2	Maintenance and repair	N.A.
4.15	ENVIRONMENTAL LOAD	
4.15.1	Wastewater treatment process	N.A.
4.15.2	Concentrations	N.A.
4.15.3	Appearance of flue gas discharge - any mist problems?	None

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
4.16	FGD SYSTEM OPERATION	
4.16.1	% of boiler load FGD can operate	Any load having gas temp. above 350°C
4.16.2	Lag time	Immediately recovery takes an hour to reach optimum efficiency.
5.	ACTUAL COSTS FOR TOTAL FGD INSTALLATION, year	1973
5.1	CAPITAL COSTS, $	
5.1.1	Scrubber	N.A.
5.1.2	Absorbent/regeneration system	N.A.
5.1.3	Sludge disposal system	N.A.
5.1.4	Site improvements	N.A.
5.1.5	Land, roads, tracks, substation	N.A.
5.1.6	Engineering costs	N.A.
5.1.7	Contractor's fee	N.A.
5.1.8	Interest during construction	N.A.
	Construction period, months	N.A.
5.1.9	License fees	N.A.
5.1.10	Other	N.A.
5.1.11	Total, $	About 3.3 million 82.5

(continued)

APPENDIX A (continued)

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
5.2	ANNUAL COSTS, $	N.A
5.2.1	Interest	
5.2.2	Depreciation	
5.2.3	Insurance and taxes	
5.2.4	Total fixed costs	
5.2.5	Labor and overhead	
	Average man-hour cost cost/year	
5.2.6	Raw material breakdown Utility cost Annual cost	
5.2.7	Utilities, unit cost, annual costs	
	Electricity Water Steam Fuel	
5.2.8	Maintenance cost/year	
5.2.9	Other costs, list	
5.2.10	Total variable cost/year	
5.2.11	Total annual cost	
5.3	WASTE DISPOSAL COST, $	N.A.
	Unit cost	
	Annual cost	
5.4	BY-PRODUCT CREDIT, $	N.A.
5.4.1	By-product 1	
5.4.2	By-product 2	

(continued)

		NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
5.5	NET ANNUAL COST	
5.5.1	For last year, mills/kWh	
5.5.2	At maximum continuous rating, mills/kWh	
5.5.3	Without FGD, mills/kWh	
6.	ENERGY REQUIREMENTS, kW	
	Pumps	140
	Fan	730
	Reheat	
	Reductant chemicals	
	Other	
	Total,	870
	Per Mg of SO_2 removed, kWh/ton SO_2	
	Per kW of electricity	0.024 kW

(continued)

	NIHON SHELL GIJUTSU/SIPM NGC CORP. Yokkaichi Refinery
7. PERFORMANCE HISTORY: OPERATING PROBLEMS AND SOLUTIONS	
7.1 Scrubber system	Dust tends to deposit on the ends of cell at the inlet of the reactor system. Steam blow facilities were recently installed to remedy this.
7.2 Demister	N.A.
7.3 Reheater	N.A.
7.4 Fan	N.A.
7.5 Sludge system	N.A.
7.6 Regeneration system	N.A.
7.7 By-product system	N.A.
7.8 Other	Plugging of waste heat boiler and corrosion of quench column. Those problems are expected to be solved by changing the material and structure of those parts.
7.9 Availability	The plant has been operated intermittently to find ways to solve the problems.

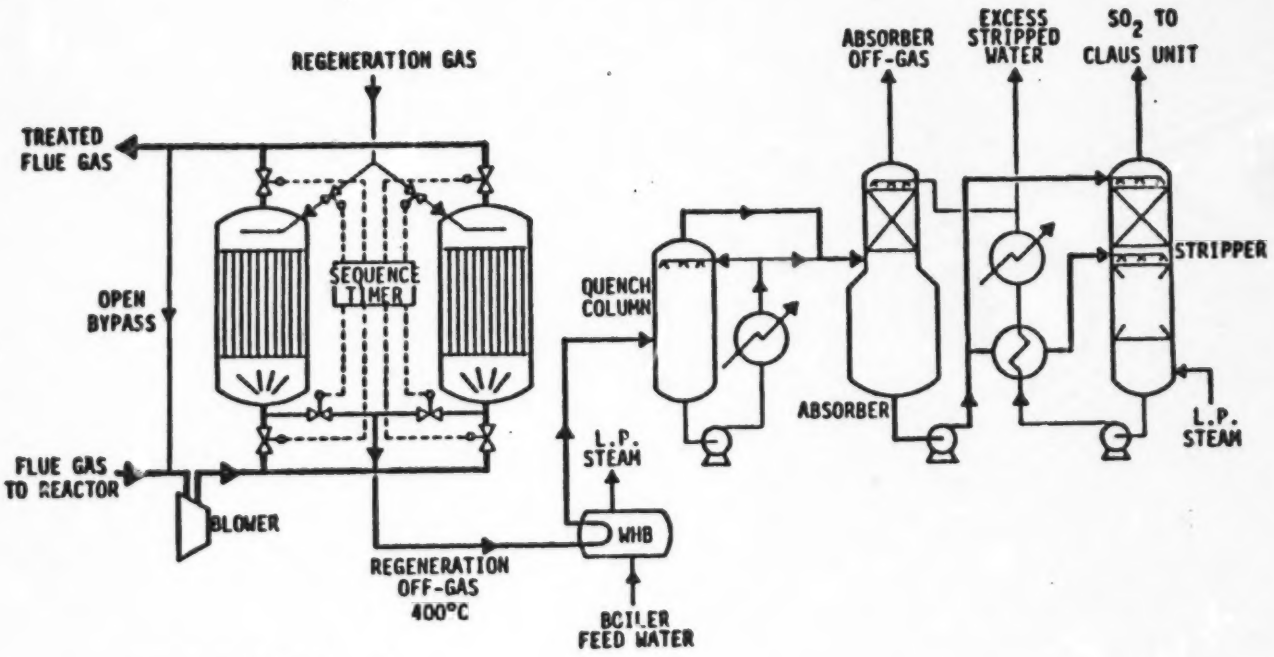

Figure A. Flowsheet of Shell process (Yokkaichi plant, SYS)

APPENDIX B

APPENDIX B

Availability Factor Computation

1. Divide boiler capacity by the number of modules and obtain MW/module = x

2. Multiply boiler capacity by number of hours during selected period (MW) = a

3. Add all down times due to module trouble for all modules during selected period = b

4. Add all down times due to boiler trouble or reduction in electricity demand for all modules during period = c

5. Availability factor = $\frac{[a - x\ (b + c)]\ 100}{a - x\ c}$ = %

APPENDIX C

APPENDIX C. CONVERSION FACTORS USED IN THIS STUDY

Multiply	By	To Obtain
ft	x 0.30480	m
ft^3	x 28.317	liter
Btu/lb	x 0.55554	kcal/kg
	x 2.32593	kJ/kg
Btu/h	x $0.29300 \cdot 10^{-3}$	kW
acfm	x 1.6992	m^3/h
$lb/10^6$ Btu	x 1.80005	mg/kcal
	x 0.42993	mg/kJ
gal/min	x 3.78541	liter/min
	x 0.22712	m^3/h
gal/1000 ft^3	x 0.13368	liter/m^3
ft^3/min	x 1.6992	m^3/h
kcal	x 4.1868	kJ
kcal	x $1.163 \cdot 10^{-3}$	kWh
acfm	x 1.60776	Nm^3/h
gr/ft^3	x 2.288	g/m^3
gr/acfm	x 2.4182	g/Nm^3
in. H_2O	x 0.2488	kPa
in. H_2O	x 2.54	mbar
gr	x 0.0648	g
ton (short)	x 1.9072	metric ton

1 MW corresponds to 3200 to 3500 m^3/h flue gas (coal)

1 acfm = 28.317 l (60°F / 14.7 psia)
(15.5°C / 760.28 torr)

26.796 l (0°C/760 torr) = 0.026796 Nm^3

1 SO_2 = 2.927 g/liter

MW = electric power generated unless other wise specified